Stimulated supercontinuum-radiation generation of carbon disulfide by all-trans-β-carotene fluorescence enhancement effect in liquid core optical fibre*

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(Received 24 July 2009; revised manuscript received 11 March 2010)

We demonstrate stimulated supercontinuum-radiation of carbon disulfide (CS_2) influenced by biological molecules all-trans-β-carotene in liquid core optical fibre (LCOF). By virtue of the broad fluorescence characteristics and large third-order optical nonlinearities of all-trans-β-carotene, the high-order Stokes lines of stimulated Raman scattering (SRS) and the multi-order Stokes lines of stimulated Brillouin scattering (SBS) excitated by SRS are observed at low input-laser energies. The results indicate that the fluorescence not only enhances the SRS, but also the SBS. These Stokes lines generate the SRS–SBS supercontinuum radiation (RBSR). A flat-amplitude bandwidth of 110 nm from 515 nm to 625 nm is observed when a frequency-doubled Nd:YAG laser at 532 nm with an energy of 0.86 mJ is used. This result is expected to be useful for the multi-wavelength fibre laser.

Keywords: stimulated Raman scattering, stimulated Brillouin scattering, all-trans-β-carotene, liquid core optical fibre

PACC: 4265B

1. Introduction

Multi-wavelength fibre lasers have been a topic of extensive study in the past decade, because of their potential applications in optical communications, sensors, and terahertz frequency generation.[1−3] A variety of approaches to these applications that involve different materials, phenomena and Brillouin fibre lasers have not been considered seriously because of the less efficiency of these lasers for the generation of multiple lines. The application of multi-wavelength lasers is limited because of the difficulty in controlling lasing bandwidth, energy, and complex optimization.[4] Stimulated Raman scattering (SRS) is an effective way to obtain multi-wavelength light sources in silica fibres. Generally, the Raman efficiencies for silica-core fibres are usually small.[5] Recently, the Raman–Brillouin comb broadband spectrum was widely studied.[4−7] The results have enriched the physics content of the coherent light radiation and widened the range of wavelength of the stimulated radiation. However, the thresholds of both SRS and stimulated Brillouin scattering (SBS) are so high, which limit the wide applications of both SRS and SBS. In order to lower the threshold and enhance the intensity, researchers applied the fluorescent dye to enhance the SRS of a weak-gain Raman mode in a liquid droplet.[8−10] Liquid core optical fibre (LCOF) can increase the intensity of the stimulated radiation and lower the threshold of the stimulated radiation. Thus, the technique of the fluorescence seeding enhanced SRS in the liquid droplet and the technique of LCOF are used together in this study to reduce the threshold of SRS. Biological molecule all-trans-β-carotene has broad fluorescence characteristics and a very broad fluorescent band (380–800 nm). Although their fluorescence productivity is lower than that of fluorescent dye, the anticipated fluorescence intensity can be obtained through increasing the concentration of solution in a certain range. With the increase of the number of particles, the interaction between the Stokes lines and the fluorescence by laser excitation is intensified. All-trans-β-carotene has a long-chain structure of 11 π–π^* conjugated double bonds,

*Project supported by the National Natural Science Foundation of China (Grant Nos. 10774057 and 10974067).
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which makes its third-order optical nonlinearity very strong[11] that can enhance the gain factor of the stimulated liquid.[12] These characteristics make it appropriate for a core medium of LCOF. Therefore, we use the solution of all-trans-β-carotene in CS_2 as the core medium to study the supercontinuum spectrum of fluorescence enhancement stimulated radiation.

In this paper, we propose a theory of the multi-wavelength Raman–Brillouim supercontinuum radiation (RBSR), which is a demonstration of an RBSR in the fluorescence enhanced SRS and SBS-a totally distinct approach. By the use of the fluorescence enhanced SRS and SBS in LCOF, we demonstrate the possibility of supercontinuum-radiation, equal-amplitude cascaded RBSR operation with injecting fluorescence seeding all-trans-β-carotene. RBSR permits a longer interaction length and excellent stability in the nonlinear processes.[4]

2. Experiment

The all-trans-β-carotene is dissolved into the CS_2 to form the solutions, whose concentration ranges from 10^{-12} mol/L to 10^{-7} mol/L or neat CS_2. Then the solutions are injected into the hollow core optical fibres with 200-μm inter diameter, 400-μm outer diameter, and 100-cm length. The fabrication of the liquid core optical fibre and the experimental setup schematically are shown in Fig. 1.

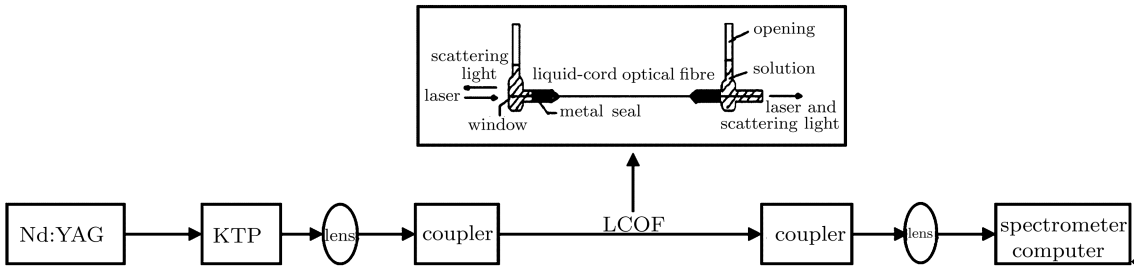


Fig. 1. Experimental setup.

As an excitation laser, a frequency-doubled Nd:YAG laser is used with a pulse duration of 30 ns, repetition rate of 10 Hz at 532-nm laser, which lies in an electronic absorption band of the all-trans-β-carotene.[13] The output light from LCOF is focused on a spectrometer by a lens; the signal is processed by a computer. We observe the high-order Raman and Brillouin Stokes lines of the all-trans-β-carotene solutions and the Raman Stokes lines of the neat CS_2 solution. Finally, the relationship between the fluorescence profile of all-trans-β-carotene and the RBSR is analysed.

3. Experimental result and discussion

Some researchers studied the fluorescent dye enhanced SRS in the liquid droplet[9−11] and obtained good results. We study the fluorescence seeding enhanced SRS in LCOF, where the intensity of Stokes lines is increased and the threshold of SRS is drastically lowered. Simultaneously, the experimental results show that the fluorescence seeding enhances not only the SRS, but also the SBS. The second-order Stokes lines of SRS and the multi-order Stokes lines of SBS are observed from an all-trans-β-carotene solution of 10^{-7} mol/L by using a pump laser with 0.12-mJ pulse energy. Without fluorescence seeding, when the pulse energy is 1.13 mJ, only the second-order Stokes lines of SRS can be observed in Figs. 2(a) and 2(b). The thresholds of different order Stokes lines in an all-trans-β-carotene solution with a concentration of 10^{-7} mol/L and in a pure CS_2 are shown in Table 1 separately.

The Brilliouin gain factor and the acoustic phonon lifetime are $g_{\rm B} = 2k\gamma^2\tau/cn\rho_0\nu$ and $\tau = \rho_0/k_{\rm s}\eta$, respectively.[14] The separation between of molecules is very short, the mass density ρ_0 and the viscosity coefficient η are big in liquid. We can obtain $g_{\rm B} \propto \tau$, $\tau \propto \rho_0/\eta$, so the different concentrations of solutions have different mass densities and viscosity coefficients. In this experiment, when the concentration is 10^{-7} mol/L, $g_{\rm B}$ is an optimal value for the supercontinuum radiation.

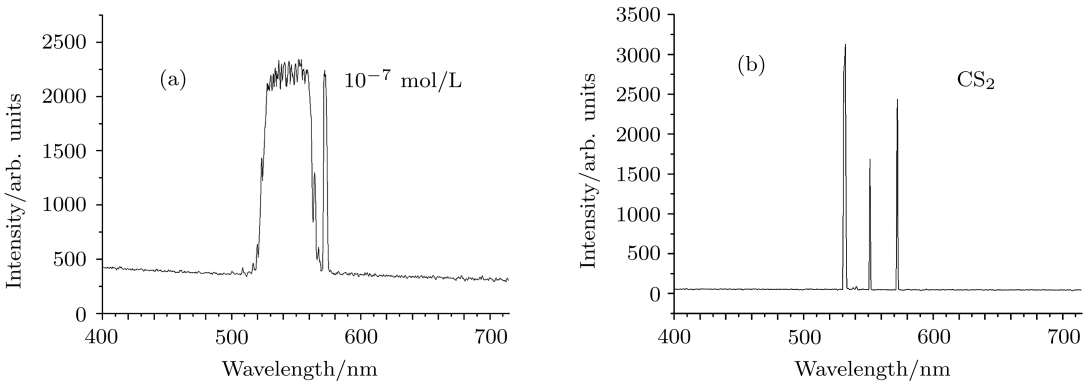


Fig. 2. The second-order Stokes line of SRS and the multi-order Stokes lines of SBS are observed at an energy of 0.12 mJ (a). The core medium is a pure CS_2 solution, the first and second-order Stokes lines are only observed with an energy of 1.13 mJ (b). The laser has a 532-nm wavelength, 30-ns pulse duration and 10-Hz repetition rate. The hollow core optical fibres each have a core diameter of 200 μm and an outer diameter of 400 μm and a length of 100 cm, and the concentration of the solution of all-trans-β-carotene in CS_2 is 10^{-7} mol/L.

Table 1. Thresholds (in mJ) of different order Stokes lines in different solutions.

solution	order number						
	1	2	3	4	5	6	7
all-trans-β-carotene in CS_2	0.06	0.12	0.15	0.21	0.41	0.50	0.86
pure CS_2	0.71	1.13	1.37	1.55	1.79	2.26	

SRS and SBS belong to the third-order nonlinear optical effect, in the standard treatment of SRS and SBS without external seeding, the exponential building of Stokes light in an optical cell is initiated by quantum noise. The SRS and the SBS intensities are governed by the following equations respectively:

$$I_{\rm RS}(L) = I_{\rm RN}(0)\left\{\exp\left[\frac{g_{\rm R}I_0}{\alpha_{\rm p}}(1-\exp(-\alpha_{\rm p}L))-\alpha_{\rm S}L\right]\right\}, \tag{1}$$

$$I_{\rm BS}(0) = I_{\rm BN}(L)\left\{\exp\left[\frac{g_{\rm B}I_0}{\alpha_{\rm p}}(1-\exp(-\alpha_{\rm p}L))-\alpha_{\rm S}L\right]\right\}, \tag{2}$$

where $I_{\rm RS}$ is the Stokes intensity of SRS, $I_{\rm BS}$ is the Stokes intensity of SBS, I_0 is the pump laser intensity. $I_{\rm RN}$ is the spontaneous noise intensity in the Raman gain bandwidth, $I_{\rm BN}$ is the spontaneous noise intensity in the Brillouin gain bandwidth, $g_{\rm R}$ is the Raman gain factor at the Stokes shift, $g_{\rm B}$ is the Brillouin gain factor at the Stokes shift, L is the length of interaction between the pump light and the Raman or Brillouin scattering medium, $\alpha_{\rm s}$ and $\alpha_{\rm p}$ are the loss coefficients of the Stokes light and the pump laser respectively. With internal seeding by fluorescence, the Stokes radiation no longer needs to start only from the spontaneous noise but can also be initiated from the fluorescence in Raman or Brillouin gain bandwidth. Hence,

$$I_{\rm RS}(L) = (I_{\rm RN}(0)+I_{\rm f}(\omega_{\rm RS})\left\{\exp\left[\frac{g_{\rm R}I_0}{\alpha_{\rm P}}(1-\exp(-\alpha_{\rm P}L))-\alpha_{\rm s}L\right]\right\}, \tag{3}$$

$$I_{\rm BS}(0) = (I_{\rm BN}(L)+I_{\rm f}(\omega_{\rm BS})\left\{\exp\left[\frac{g_{\rm B}I_0}{\alpha_{\rm P}}(1-\exp(-\alpha_{\rm P}L))-\alpha_{\rm s}L\right]\right\}, \tag{4}$$

where $I_{\rm f}(\omega_{\rm RS})$ and $I_{\rm f}(\omega_{\rm BS})$ are the fluorescence intensities at frequencies $\omega_{\rm RS}$ and $\omega_{\rm BS}$; $\omega_{\rm RS}$ and $\omega_{\rm BS}$ are the Stokes frequencies of SRS and SBS respectively. The fluorescence intensity is proportional to the product of the fluorescence cross section and the number of fluorescence dye molecules. $I_{\rm f} > I_{\rm SN}$ or $I_{\rm BN}$ even at a very low dye concentration, since the fluorescence cross section is much larger than the Raman cross section.[8] According to Eqs. (3) and (4), the thresholds of SRS and SBS can be effectively lowered by the dye-fluorescence.

About the generation of high-order Stokes of

SRS and SBS, the broadband fluorescence is another important factor. Biological molecule all-trans-β-carotene with the broad fluorescence characteristics, and its fluorescence bandwidth is so broad that the multi-order Stokes lines of SRS are easy to obtain.

The seventh-order Stokes line of SRS can be observed when the input-laser energy is 0.86 mJ. The threshold increments of Stokes lines are correlative with the fluorescence profile of all-trans-β-carotene: when the fluorescence intensity in the wavelength region of the spectrum is high, the threshold increment of Stokes line is small, and *vice versa* as shown in Ref. [15]. The threshold of SRS depends on fluorescence intensity: the stronger the fluorescence intensity is, the lower the threshold increment will be; and *vice versa*. The threshold increment is the difference in threshold between adjacent Stokes. The threshold increment of the fifth-order Stokes line drastically increases at the different concentrations because the fifth-order Stokes wavelength is located at the place where the fluorescence intensity is weak. When the experimental energy ranges from 0 to 5 mJ and the wavelength is located at the place where the fluorescence intensity is weak, the higher-order Stokes lines cannot be observed.

According to the Raman threshold definition

$$I_{\rm s}(L) = I_{\rm p}(L) = I_0 \exp(-\alpha_{\rm p} L), \tag{5}$$

where $I_{\rm p}$ is the pump light intensity from the output end of the fibre. When the length of the LCOF is 100 cm, it can be proved that $g_{\rm R} = 10^{-9}$ cm/W, and $I_{\rm RN}(0) = 0.001$ W/cm^2.[16] According to the frequency of light and the concentration of all-trans-β-carotene, $I_{\rm f}(\omega_{\rm RS})$ ranges from 0.008 W/cm^2 to 0.013 W/cm^2.[8] CS_2 has a less light absorption loss. In our experiment, $\alpha_{\rm s} = \alpha_{\rm p} = 0.003$ cm^{-1}.[17] Therefore, the thresholds at different concentrations can be obtained after simultaneous Eqs. (3) and (5) have been solved. For example, at a concentration of 10^{-7} mol/L, we obtain the value of $I_{\rm f}(\omega_{\rm RS})$ to be 0.008–0.011 W/cm^2 according to different order numbers (1–7 orders). The fitting curve is shown in Fig. 3, which is well consistent with the experimental results.

Owing to the duple effects of broadband fluorescence and LCOF, SRS and SBS of the stimulated liquid are simultaneously excited, when the pulse duration, the geometric parameter of LCOF, the concentration of the fluorescent matters and the stimulated liquid characteristics are consistent with those required by the experimental conditions. In this experiment, the SBSR is composed of the multi-order SRS and the multi-order SBS that is excited by the SRS, and can be detected at a lower input-energy. The stimulated radiation whose bandwidth is approximately 110 nm, can be observed and shown in Fig. 4.

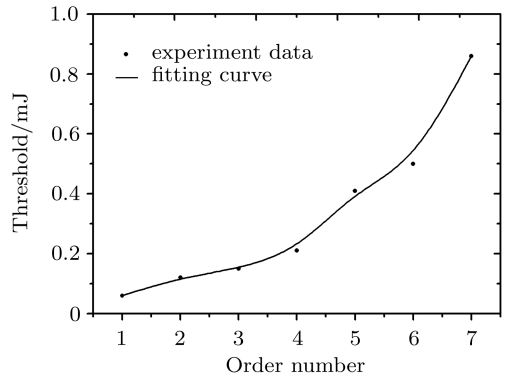


Fig. 3. Threshold numerical simulation curve of 1–7 order number Stokes lines. The solid curve is the fitting curve, and the solid circles denote the experimental data. The concentration is 10^{-7} mol/L.

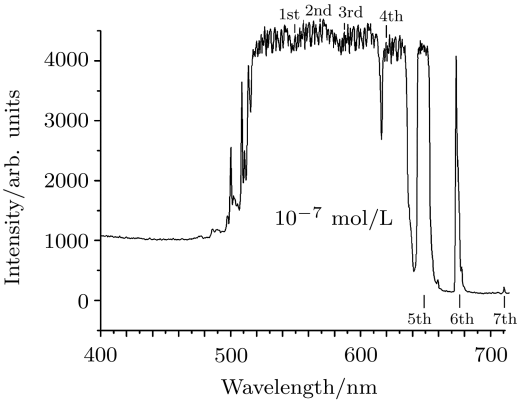


Fig. 4. SBSR spectrum, which is composed of the seventh-order SRS and the multi-order SBS that is excitated by SRS. When the laser has 532-nm wavelength, 0.86-mJ energy, 30-ns pulse duration, 10-Hz repletion rate, and the hollow core optical fibres each have a core diameter of 200 μm and an outer diameter of 400 μm and a length of 100 cm, the concentration of solution is 10^{-7} mol/L.

4. Conclusion

Fluorescence seeding all-trans-β-carotene enhances not only the SRS, but also the SBS. Owing to the duple enhancement effects of the broadband fluorescence and the LCOF, the Raman–Brillouin supercontinuum radiation can be obtained at a low input-laser energy. We believe that this result might have a wide range of applications for broadband fibre laser, seeding laser and optical amplification.

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